On Silicon Carbide Grains as the Carrier of the 21 Micron Emission Feature in Post-Asymptotic Giant Branch Stars

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ABSTRACT

The mysterious $21\,\mu$m emission feature seen in 12 proto-planetary nebulae (PPNe) remains unidentified since its first detection in 1989. Over a dozen of candidate materials have been proposed within the past decade, but none of them has received general acceptance. Very recently, silicon carbide (SiC) grains with impurities were suggested to be the carrier of this enigmatic feature, based on recent laboratory data that doped SiC grains exhibit a resonance at $\sim 21\,\mu$m. This proposal gains strength from the fact that SiC is a common dust species in carbon-rich circumstellar envelopes. However, SiC dust has a strong vibrational band at $\sim 11.3\,\mu$m. We show in this *Letter* that in order to be consistent with the observed flux ratios of the $11.3\,\mu$m feature to the $21\,\mu$m feature, the band strength of the $21\,\mu$m resonance has to be very strong, too strong to be consistent with current laboratory measurements. But this does not yet readily rule out the SiC hypothesis since recent experimental results have demonstrated that the $21\,\mu$m resonance of doped SiC becomes stronger as the C impurity increases. Further laboratory measurements of SiC dust with high fractions of C impurity are urgently needed to test the hypothesis of SiC as the carrier of the $21\,\mu$m feature.

Subject headings: circumstellar matter — dust, extinction — infrared: stars — stars: AGB and Post-AGB — stars: individual (HD 56126)

1. Introduction

The so-called "$21\,\mu$m feature" has been identified in 12 proto-planetary nebulae (PPNe; Kwok, Volk, & Hrivnak 1999) (and arguably also in in 2 planetary nebulae [PNe] associated

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with Wolf-Rayel central stars [Hony, Waters, & Tielens 2001] and in 2 highly evolved carbon stars [Volk, Xiong, & Kwok 2000]) since its first detection in 1989 (Kwok et al. 1989). This feature has a similar spectral shape and peaks at the same wavelength ($\sim 20.1\,\mu$m) in all sources. The 21 μm-feature sources have quite uniform properties: they are mostly metal-poor, carbon-rich F and G supergiants with infrared excesses and overabundant s-process elements (see Kwok et al. 1999).

The origin of this feature, however, is still a mystery. A large number of candidate carriers have been proposed in the past decade, including hydrogenated fullerenes, polycyclic aromatic hydrocarbon, hydrogenated amorphous carbon, diamonds, synthetic carbonaceous macromolecules, amides (thiourea or urea $OC\,[NH_2]_2$), iron oxides (such as Fe_2O_3 or Fe_3O_4), SiS_2, oxygen-bearing side groups in coal (see Kwok et al. 1999, Andersen, Posch, & Mutschke 2005 and references therein), and more recently titanium carbide (TiC) nanoclusters (von Helden et al. 2000), stochastically-heated silicon core-SiO_2 mantle nanograins (Smith & Witt 2002; Li & Draine 2002), doped SiC (Speck & Hofmeister 2004), SiC core-SiO_2 mantle grains and iron monoxide FeO (Posch, Mutschke, & Andersen 2004).

Among these candidates, TiC nanograins have recently received much attention, because (1) laboratory spectra of TiC nano-crystals exhibits a distinct feature at $\sim 20.1\,\mu$m, closely resembling the astronomical 21 μm emission feature both in peak position and width and in spectral details (von Helden et al. 2000), although bulk TiC does not show any noticeable feature near 20.1 μm (Henning & Mutschke 2000); (2) presolar TiC grains are identified in primitive meteorites as nanometer-sized inclusions embedded in micrometer-sized presolar graphite grains (Bernatowicz et al. 1996). However, the TiC model has been challenged by Hony et al. (2003), Chigai et al. (2003), and Li (2003).[1]

More recently, SiC seems to be an attractive candidate for the 21 μm feature carrier: Speck & Hofmeister (2004) reported the experimental finding that SiC, under certain circum-

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[1] Hony et al. (2003) found that nano TiC must absorb much more strongly in the optical and ultraviolet (UV) wavelengths than its bulk counterpart in order for the required amount of TiC dust not to exceed the (observed) maximum available Ti abundance. Chigai et al. (2003) found that in order to be consistent with the observed flux ratio of the 21 μm and 11.3 μm bands, the Ti/Si abundance ratio must be at least 5 times larger than the solar abundance ratio. One may argue that the arguments of Hony et al. (2003) and Chigai et al. (2003) do not readily rule out the TiC hypothesis because (1) as a consequence of the so-called *electron mean free path limitation* effect, the imaginary parts of the dielectric functions of small metallic grains (and therefore their optical/UV absorptivities) are expected to be larger than those of their bulk counterparts (see Li 2004); (2) there is no reason to compare the solar Ti/Si abundance ratio with that of the 21 μm-feature sources. However, applying the Kramers-Kronig physical principle to the TiC hypothesis, Li (2003) readily ruled out the TiC model because it was found that this model requires at least 50 times more TiC mass than available, no matter how strong the UV/optical absorptivity of nano TiC is.

stances, not only shows the well-known resonance feature at $\sim 11.3\,\mu$m, but also a secondary band which is centered at 20–21 μm. This secondary band was reported to appear only in the β SiC-polytype, and nitrogen or carbon impurities were suspected to favor its occurrence. This hypothesis gains strength from the fact that (1) both silicon and carbon are abundant elements; and (2) SiC is a common dust species in C-rich circumstellar envelopes. Due to the astrophysical significance of SiC dust, it deserves a more thorough investigation. In this *Letter*, we aim at investigating whether doped-SiC can be a suitable carrier for the mysterious 21 μm feature by comparing the model-predicted flux ratio of the 21 μm feature to the 11.3 μm feature with observed.

2. Circumstellar SiC Grains and Their Optical Properties

As early as 1933 Wildt had already suggested that SiC grains might form in the cool atmospheres of N-type stars. This suggestion was confirmed 36 years later when Gilman (1969) and Friedemann (1969) performed thermodynamical equilibrium calculations and found that SiC grains could condense in carbon stars. The presence of circumstellar SiC grains was first revealed by the detection of an emission feature at 11.5 μm in some carbon stars which was first attributed to SiC dust by Gilra (1972). The detection of this feature and its identification as SiC were confirmed by subsequent observations and interpretations (Treffers & Cohen 1974; Merrill & Stein 1976; Goebel et al. 1980; Little-Marenin 1986; Goebel, Chesseman, & Gerbaut 1995; Speck, Barlow, & Skinner 1997). The formation of SiC dust in carbon stars has also been indicated by the identification of presolar SiC grains in primitive meteorites based on their isotopic anomalies (Bernatowicz et al. 1987).

SiC has ~ 70 polytypes, which can be divided into 2 general crystallographic types: cubic (β SiC) and hexagonal (α SiC). While presolar SiC grains were predominantly found to have a cubic lattice structure (i.e. β SiC; Daulton et al. 2003), the 11.3 μm emission feature observed for carbon-rich AGB stars is best fitted by laboratory spectra of α SiC (Baron et al. 1987). Speck, Hofmeister, & Barlow (1999) suggested that this discrepancy (between meteoritic and astronomical identifications of the SiC type) is caused by the "inappropriate 'KBr corrections' made to laboratory spectra of SiC taken using the KBr matrix method"; they argued that the carrier of the 11.3 μm feature seen in carbon stars is actually β SiC (instead of α SiC). More recently, Clément et al. (2003) found that the laboratory spectra of matrix-isolated β SiC nanoparticles perfectly match the astronomical 11.3 μm feature. But Mutschke et al. (1999) argued that the 11.3 μm emission feature itself is not a powerful discriminator of the SiC crystal type (also see Papoular et al. 1998).

Therefore, in this work we will consider both two types of SiC grains. We approximate

the grains as spherical[2] and use Mie theory to calculate their absorption and scattering properties. The complex index of refraction $m(\lambda) = m'(\lambda) + i\, m''(\lambda)$ of α SiC is taken from Laor & Draine (1993).[3] For β SiC, Adachi (1999) compiled the refractive indices recently determined by various groups in the wavelength range of $0.13\,\mu\mathrm{m} \le \lambda \le 124\,\mu\mathrm{m}$, except there was no m'' data for $0.5\,\mu\mathrm{m} \le \lambda \le 0.65\,\mu\mathrm{m}$. Largely based on the data compiled by Adachi (1999), we take the following "synthetic" approach: for $\lambda \le 0.13\,\mu\mathrm{m}$, we take the imaginary parts of the refractive indices m'' from Laor & Draine (1993);[4] for $0.13\,\mu\mathrm{m} \le \lambda \le 0.5\,\mu\mathrm{m}$ and $0.65\,\mu\mathrm{m} \le \lambda \le 124\,\mu\mathrm{m}$, we take m'' from Adachi (1999); for $0.5\,\mu\mathrm{m} \le \lambda \le 0.65\,\mu\mathrm{m}$, extrapolation is made from that of Adachi (1999) at $\lambda \le 0.5\,\mu\mathrm{m}$; for $\lambda > 124\,\mu\mathrm{m}$, we assume $m''(\lambda) = m''(124\mu\mathrm{m})(124\mu\mathrm{m}/\lambda)$. After smoothly joining the adopted m'', we calculate the real part $m'(\lambda)$ from m'' through the Kramers-Kronig relation (Bohren & Huffman 1983).

If SiC grains are indeed the carrier of the observed $21\,\mu\mathrm{m}$ feature, they must have a resonance at this wavelength and we should include its contribution to their dielectric functions. We approximate this contribution in terms of a single Lorentz oscillator

$$\delta\epsilon = \frac{\omega_{\mathrm{p}}^2}{\omega_0^2 - \omega^2 - i\,\gamma\omega} \tag{1}$$

where $\omega \equiv 2\pi c/\lambda$ is the angular frequency (c is the speed of light), ω_0 is the angular frequency of the transverse optical mode, $\gamma = 2\pi c\Delta\lambda_0/\lambda_0^2$ is the damping constant ($\lambda_0 \approx 20.1\,\mu\mathrm{m}$ and $\Delta\lambda_0 \approx 2\,\mu\mathrm{m}$ are respectively the peak wavelength and the FWHM of the $21\,\mu\mathrm{m}$ feature), and ω_{p} is the plasma frequency. For spherical grains in the Rayleigh regime, the parameters ω_{p} and ω_0 can be determined from the feature strength (Q_{abs}/a) (Bohren & Huffman 1983)[5]

$$\omega_{\mathrm{p}} = \frac{1}{3}\left[\left(\frac{Q_{\mathrm{abs}}}{a}\right)\frac{3\gamma c}{4}\right]^{1/2}\left[\epsilon(\infty)+2\right] \quad , \tag{2}$$

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[2]If the grain shape is approximated as a continuous distribution of ellipsoids (Bohren & Huffman 1983), both the $11.3\,\mu\mathrm{m}$ feature and the $21\,\mu\mathrm{m}$ feature will be significantly broadened. But since at these wavelength ranges micron or submicron-sized SiC grains are in the Rayleigh regime, the broadening effect is unlikely to differ much from one feature to another. Therefore, it is sufficient to just consider spherical grains.

[3]Laor & Draine (1983) based their dielectric functions ($\epsilon = m^2$) in the $11.3\,\mu\mathrm{m}$ wavelength range on those of Spitzer, Kleinman, & Walsh (1959), but broadened by a factor of ~ 16.7. In addition, they introduced a "continuum" by adding a highly damped oscillator.

[4]At such short wavelengths, the dielectric functions for α SiC and β SiC should not differ much since they just depend on the atomic absorption cross sections of the constituent atoms and they are not sensitive to the exact crystal structure.

[5]Q_{abs} is the absorption efficiency at $\lambda_0 = 20.1\,\mu\mathrm{m}$ and a is the grain radius. For grains in the Rayleigh approximation (i.e. $2\pi a/\lambda \ll 1$), $(Q_{\mathrm{abs}}/a) \equiv (4\rho/3)\,\kappa_{\mathrm{abs}}$ is independent of a, where $\rho \approx 3.22\,\mathrm{g\,cm^{-3}}$ is the mass density of SiC and κ_{abs} is the mass absorption coefficient of SiC at $20.1\,\mu\mathrm{m}$. In this work, Q_{abs}/a is treated as a free parameter.

$$\omega_0 = \left[\left(\frac{2\pi c}{\lambda_0}\right)^2 - \frac{\omega_{\rm p}^2}{\epsilon(\infty)+2}\right]^{1/2} \tag{3}$$

where $\epsilon(\infty)$ is the dielectric function at $\omega \rightarrow \infty$: $\epsilon(\infty) \approx 6.6(6.7)$ for $\alpha\,$SiC ($\beta\,$SiC).

Unfortunately, Speck & Hofmeister (2004) did not measure the absolute strength of the $21\,\mu$m feature ($Q_{\rm abs}/a$) for their SiC samples. We therefore treat ($Q_{\rm abs}/a$) as a free parameter: for a given ($Q_{\rm abs}/a$) value, we calculate the contribution of the $21\,\mu$m feature to the dielectric functions of SiC from eqs.(1,2,3) and add this component to the dielectric functions of Laor & Draine (1993) for $\alpha\,$SiC and those described early in this section for $\beta\,$SiC.[6] For illustrative purpose, in Figure 1 we plot the refractive indices of $\alpha\,$SiC and $\beta\,$SiC with $(Q_{\rm abs}/a) = 0, 100, 10^3, 10^4\,{\rm cm}^{-1}$.

3. Results

The $21\,\mu$m sources all have a weak emission feature at $11.3\,\mu$m, which is commonly attributed to PAHs (see Kwok et al. 1999). Assuming that all the power from the $11.3\,\mu$m feature is emitted by SiC grains, one can place constraints on the size and the $21\,\mu$m feature strength ($Q_{\rm abs}/a$) of SiC dust by comparing the power emitted from the $11.3\,\mu$m feature with that emitted from the $21\,\mu$m feature. For this purpose, we take HD 56126 (for which the dust and gas spatial distributions are well constrained; see Hony et al. 2003, Meixner et al. 2004, Hrivnak & Bieging 2005) as a testing example.

HD 56126 (IRAS 07134+1005), a bright post-AGB star with a spectral type of F0-5I, is one of the four $21\,\mu$m sources originally discovered by Kwok et al. (1989) and remains the best-studied $21\,\mu$m source. The total power emitted from the $11.3\,\mu$m feature and from the $21\,\mu$m feature are respectively $E(11.3\mu{\rm m}) \approx 1.8 \times 10^{-11}\,{\rm erg\,s^{-1}\,cm^{-2}}$ and $E(21\mu{\rm m}) \approx 1.5 \times 10^{-9}\,{\rm erg\,s^{-1}\,cm^{-2}}$ (Hony et al. 2003). Now the question is, with what grain size and what ($Q_{\rm abs}/a$) for the $21\,\mu$m feature one can achieve $E(11.3\mu{\rm m})/E(21\mu{\rm m}) < 0.012$?

Apparently, the observational requirement of $E(11.3\mu{\rm m})/E(21\mu{\rm m}) < 0.012$ is best met if the grains are cold (say, with an equilibrium temperature $\sim 150\,$K) and have a large $21\,\mu$m feature strength ($Q_{\rm abs}/a$). Since the further the grains are away from the central illuminating star, the colder the grains are, we just need to consider how cold SiC dust can be at the outer edge of the dusty envelope around HD 56126. On the other hand, ($Q_{\rm abs}/a$) for the

[6] The Lorentz oscillator nature of the $21\,\mu$m resonance (see eq.[1]) guarantees that the new dielectric functions also satisfy the Kramers-Kronig relation (see Bohren & Huffman 1983).

21 μm feature can not be arbitrarily large – it should not be inconsistent with laboratory measurements (e.g. see Fig. 1 in Speck & Hofmeister 2004).

The equilibrium temperature of a SiC grain of spherical radius a at the the outer edge of the dust envelope around HD 56126 can be determined by balancing absorption and emission

$$\left(\frac{R_\star}{2r_{\rm max}}\right)^2 \int_0^\infty C_{\rm abs}(a,\lambda) F_\lambda^\star \exp(-A_\lambda/1.086) d\lambda = \int_0^\infty C_{\rm abs}(a,\lambda) 4\pi B_\lambda\left(T[a]\right) d\lambda \qquad (4)$$

where $R_\star \approx 49.2\, r_\odot$ is the stellar radius ($r_\odot$ is the solar radius); $r_{\rm max} \approx 9.3\times10^{16}$ cm is the distance from the central star to the outer edge of the dust envelope (Hony et al. 2003); $C_{\rm abs}(a,\lambda)$ is the absorption cross section of SiC dust of size a at wavelength λ; $T(a)$ is the equilibrium temperature of dust of size a at $r_{\rm max}$; $F_\lambda^\star$ is the flux per unit wavelength ($\rm erg\, s^{-1}\, cm^{-2}\, \mu m^{-1}$) at the top of the illuminating star's atmosphere which is approximated by the Kurucz (1979) model atmospheric spectrum with $T_{\rm eff} = 7250$ K and $\log g = 1.0$; and A_λ is the dust extinction which is represented by the Milky Way standard extinction law with $A_V = 1.0$ mag (Hony et al. 2003).[7] For a SiC grain of a given size a and a given 21 μm feature strength $(Q_{\rm abs}/a)$, we calculate its equilibrium temperature from eq.(4) and its emission spectrum and then calculate $E(11.3\mu{\rm m})/E(21\mu{\rm m})$ – the ratio of the amount of energy emitted from the 11.3 μm feature to that from the 21 μm feature.

In Figure 2 we plot the emission spectra of α SiC grains of sizes a = 0.01, 0.05, 0.1, 0.5 and 1.0 μm with $(Q_{\rm abs}/a) = 100, 1000, 10^4\,{\rm cm^{-1}}$ for the 21 μm feature. It is seen that even if one assumes $(Q_{\rm abs}/a) = 10^4\,{\rm cm^{-1}}$, the calculated $E(11.3\mu{\rm m})/E(21\mu{\rm m})$ ratio (≈ 0.61 for $a = 0.01\,\mu$m and ≈ 0.41 for $a = 1.0\,\mu$m) is still much larger than the observed ratio of $E(11.3\mu{\rm m})/E(21\mu{\rm m}) < 0.012$.[8] In order to be consistent with the observation, one requires $(Q_{\rm abs}/a) \gg 10^4\,{\rm cm^{-1}}$. This seems unlikely since the required strength for the 21 μm feature is stronger than that for the 11.3 μm feature of α SiC which is only $(Q_{\rm abs}/a) \approx 1.5\times10^4\,{\rm cm^{-1}}$.[9] Similar results are obtained for β SiC (see Fig. 3).[10]

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[7]This extinction correction is not exact. But it does not affect our results since we only need to approximately estimate the amount of dust attenuation (see Fig. 1 of Hony et al. 2003).

[8]Note that $E(11.3\mu{\rm m})/E(21\mu{\rm m}) < 0.012$ is already a very generous upper limit since we have attributed the entire 11.3 μm emission to SiC, while actually this emission must at least partly originate from PAHs.

[9]As a matter of fact, in the experimental spectra of the β SiC samples of Speck & Hofmeister (2004), the 21 μm resonance is far weaker than the 11.3 μm resonance for both bulk and nano materials.

[10]In addition to the prominent 11.3 μm band, β SiC appears to have another sharp band at $\sim$12.6 μm. This is caused by the asymmetrical nature of its index of refraction (Adachi 1999; see Fig. 1d). If we approximate the 11.3 μm resonance by a Lorentz oscillator, the secondary peak at $\sim$12.6 μm would disappear. But this

4. Discussion

In §3 we only consider grains large enough to attain an equilibrium temperature. For small grains with $a < 100\,\text{Å}$, they will undergo single-photon heating: upon absorption of an energetic photon, they will be heated to a temperature which is higher than their equilibrium temperature, and most of the photon energy will be radiated away at this high temperature. Therefore, the emission spectrum of a stochastically-heated grain peaks at shorter wavelengths than that calculated from its equilibrium temperature (see Draine & Li 2001). We thus would expect larger ratios of $E(11.3\mu\text{m})/E(21\mu\text{m})$ for small grains undergoing single-photon heating than those considered above, making things even worse.

However, it is still premature to rule out the SiC hypothesis since the mid-IR spectra of doped SiC samples of Speck & Hofmeister (2004) were obtained at *room temperature* for SiC with a limited fraction of C impurity. While it is unclear how and to what degree temperatures will affect the $11.3\,\mu$m and $21\,\mu$m features of SiC, it is known that the mid-IR spectra of silicates are affected by the sample temperature (See Bowey et al. 2001). Moreover, recent experimental results have shown that the strength of the $21\,\mu$m feature increases as the impurity content increases (Kimura et al. 2005a,b). The required $21\,\mu$m feature strength (see §3) might be attainable by SiC dust with a rather high level of impurity. Laboratory measurements of the mid-IR spectra of heavily doped SiC samples are urgently needed.

Finally, we note that there are over 700 C-rich AGB stars known to show the $11.3\,\mu$m SiC feature from the IRAS (*Infrared Astronomical Satellite*) LRS (*Low Resolution Spectrometer*) survey, but none of these stars show the $21\,\mu$m feature, suggesting that the amount of impurity incorporated into SiC dust must be environment dependent (e.g. the Si/C ratio, Speck & Hofmeister 2004), if doped SiC is indeed the carrier of the $21\,\mu$m feature.

In summary, we have examined the recent hypothesis of doped-SiC as the carrier of the mysterious $21\,\mu$m emission feature detected in 12 PPNe. It is found that doped SiC grains have to have a resonance at $\sim 21\,\mu$m too strong to be consistent with current laboratory measurements, in order for the model-predicted flux ratios of the $11.3\,\mu$m feature to the $21\,\mu$m feature not in conflict with the observed values. Admittedly, it is still premature to discard the SiC hypothesis since recent experimental results have shown that the strength of the $21\,\mu$m resonance of doped SiC appears to increases with the C impurity content, suggesting that heavily doped SiC may be able to produce a sufficiently strong $21\,\mu$m resonance. We call on laboratory measurements of the mid-IR spectra of SiC with high levels of C impurity.

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does not affect our conclusion since we are considering the total power emitted in these features, rather than their peak heights.

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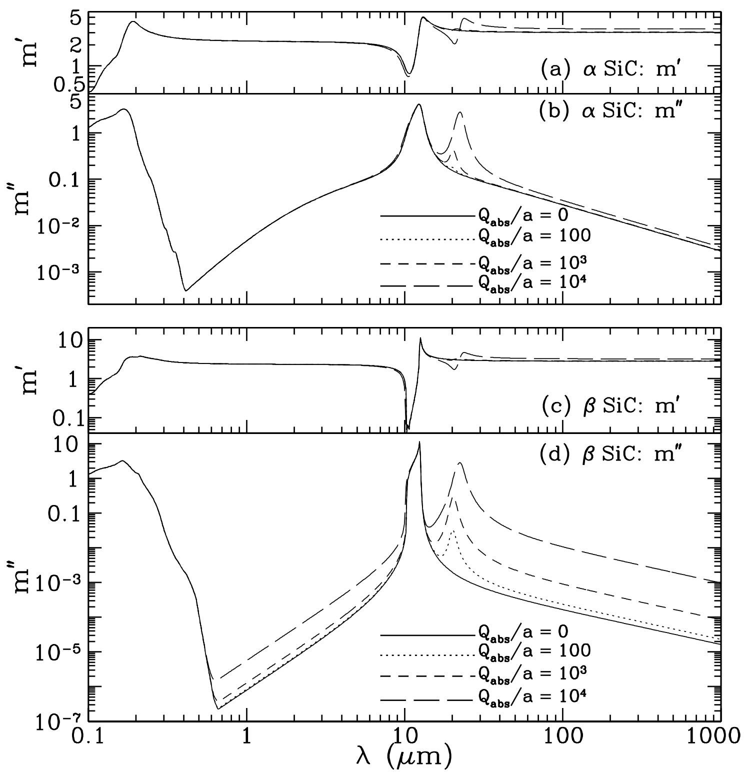

Fig. 1.— Refractive indices $m(\lambda) = m'(\lambda) + i\, m''(\lambda)$ of $\alpha\,$SiC (a,b) and $\beta\,$SiC (c,d) with various strength for the 21 μm feature: $Q_{\rm abs}/a = 0$ (solid lines), $Q_{\rm abs}/a = 100\,{\rm cm}^{-1}$ (dotted lines), $Q_{\rm abs}/a = 10^3\,{\rm cm}^{-1}$ (short-dashed lines), and $Q_{\rm abs}/a = 10^4\,{\rm cm}^{-1}$ (long-dashed lines).

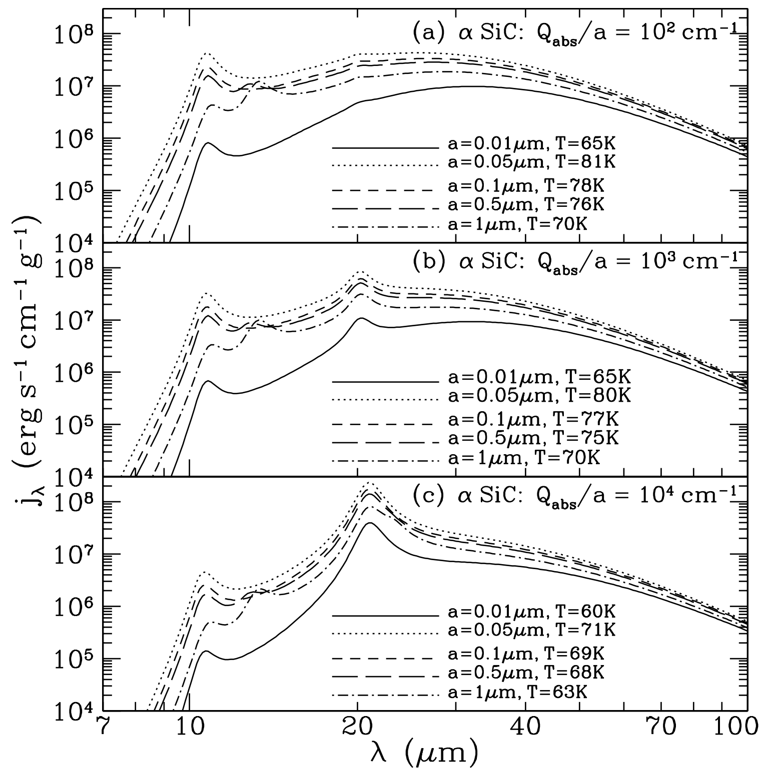

Fig. 2.— Emission spectra for $\alpha\,$SiC grains of sizes $a = 0.01\,\mu$m (solid lines), $a = 0.05\,\mu$m (dotted lines), $a = 0.1\,\mu$m (short-dashed lines), $a = 0.5\,\mu$m (long-dashed lines), and $a = 1.0\,\mu$m (dot-dashed lines), with $(Q_{\rm abs}/a) = 100\,{\rm cm}^{-1}$ (a), $(Q_{\rm abs}/a) = 10^3\,{\rm cm}^{-1}$ (b), and $(Q_{\rm abs}/a) = 10^4\,{\rm cm}^{-1}$ (c) for the $21\,\mu$m feature. The grains are at the outer edge of the dust envelope around HD 56126.

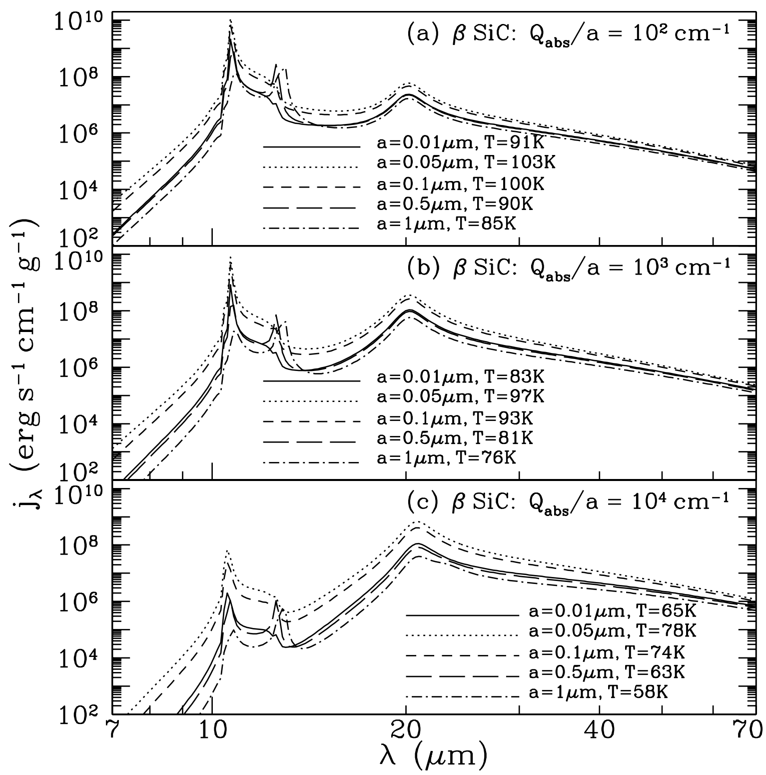

Fig. 3.— Same as Figure 2 but for β SiC.